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(54) Title: DISINFECTING COMPOSITIONS AND PROCESSES FOR DISINFECTING SURFACES

(57) Abstract

The present invention relates to the disinfection of surfaces with a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil or mixtures thereof.

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DISINFECTING COMPOSITIONS AND PROCESSES FOR DISINFECTING SURFACES

Technical field

The present invention relates to antimicrobial compositions which can be used to disinfect and clean various surfaces including animate surfaces (e.g., human skin, mouth and the like) and inanimate surfaces including, but not limited to, hard surfaces like walls, tiles, table tops, glass, bathroom surfaces, kitchen surfaces, dishes as well as fabrics, clothes, carpets and the like.

Background

Antimicrobial/antibacterial compositions include materials which have the ability to disinfect. It is generally recognised that a disinfecting material greatly reduces or even eliminates the micro-organisms, e.g., bacteria, existing on a surface. Compositions based on halogen containing compounds like hypochlorite or on quaternary compounds, have been extensively described in the art for disinfecting purpose. Compositions comprising peroxide bleach are also known as disinfecting compositions.

However, compositions comprising hydrogen peroxide as the only antimicrobial agent, typically at a level of 7% by weight of the total composition, are not fully satisfactory when used upon high dilution levels, e.g. at a dilution level of 1:50 (composition:water) to disinfect clean surfaces, i.e., surfaces which are substantially free of organic and/or inorganic residues.

It is thus a need to provide disinfecting compositions that deliver excellent disinfection on clean surfaces even when used upon high diluted conditions.

Representative of the prior art is for example WO88/00795 which discloses liquid disinfecting compositions comprising a compound selected from the group of organic acids, perborates, peracids and their salts, together with other antimicrobial compounds like a quaternary ammonium salt and an essential oil.

However, a drawback associated with liquid compositions comprising a bleaching agent and an essential oil is that said liquid compositions tend to be chemically unstable upon prolonged storage periods. Indeed, traditional essential oils are usually made by a blend of components including terpenes, esters, aromatic components, chetones, aldehydes and the like. Said essential oils are sensitive to peroxygen bleaches. In other words, said essential oils show significant instability in presence of peroxygen bleaches and tend to decompose / oxidise said peroxygen bleaches, therefore leading to a lowering of the total level of available oxygen in a composition comprising a peroxygen bleach with consequent chemical stability issues upon long storage periods.

It is therefore an object of the present invention to provide a liquid disinfecting composition comprising a peroxygen bleach which is chemically stable for long storage periods while delivering excellent disinfection performance on clean surfaces when used upon highly diluted conditions.

It has now been found that this can be achieved by providing a liquid peroxygen bleach-containing composition comprising on top of said peroxygen bleach, an antimicrobial active of essential oil or mixtures thereof. More particularly, it has been found that the liquid peroxygen bleach-containing compositions of the present invention comprising said antimicrobial active of essential oil, exhibit improved chemical stability upon long storage periods, as compared to the same compositions comprising antimicrobial essential oils instead of said antimicrobial active of essential oils, while providing also excellent disinfection on clean surfaces, i.e. surfaces being free of any organic and/or inorganic soils, even at high dilution levels, i.e. up to dilution levels of from 1:100 (composition:water).

An advantage of the present invention is that xcellent disinfection is provided on a broad range of pure bacterial strains including Gram positive and Gram negative bacterial strains and more resistant micro-organisms like fungi.

Another advantage of the compositions of the present invention is that beside the disinfection properties delivered, good cleaning is also provided, especially in the embodiment of the present invention where the compositions herein further comprise a surfactant and/or a solvent.

Also, the compositions according to the present invention are suitable to be used on all types of surfaces including animate surfaces (e.g., human skin and/or mouth when used as an oral preparation or toothpaste) and inanimate surfaces. Indeed, this technology is particularly suitable in hard-surfaces applications as well as in laundry applications, e.g., as a laundry detergent or laundry additive in a so called "soaking mode", "through the wash mode", or even as a laundry pretreater in a "pretreatment mode". More particularly, the compositions according to the present invention are suitable to be used on delicate surfaces including those surfaces in contact with food and/or babies in a safe manner. Indeed, when using the compositions according to the present invention in diluted conditions, the amount of chemical residues left onto a surface disinfected therewith is reduced. Thus, it may be not necessary to rinse for example a hard-surface after the compositions of the present invention have been applied thereto in diluted conditions.

EP-B-288 689 discloses a liquid for hard-surfaces comprising antimicrobial effective amounts of pine oil and at least one oil soluble organic acid. No other antimicrobial compounds are mentioned, let alone a peroxygen bleach.

EP-241390 discloses that textiles contaminated with bacteria may be disinfected by first treating with a detergent and then with a peroxide bleaching agent in an aqueous bath at pH 9-13 in the presence of non complexed calcium. No other antimicrobial compounds are disclosed, let alone antimicrobial actives of ssential oils.

US-4,404,191 discloses that per-compounds such as monopersulfate have bactericide, fungicide and virucide properties. US-4,404,191 discloses that compositions comprising monopersulfate may be used in diluted form to treat hard-surfaces. However no other antimicrobial compounds are disclosed, let alone antimicrobial actives of essential oils.

US-5,403,587 discloses aqueous antimicrobial compositions which can be used to sanitise, disinfect, and clean hard-surfaces. More particularly, US-5,403,587 discloses aqueous compositions (pH 1 to 12) comprising essential oils (0.02% to 5%), which exhibit antimicrobial properties efficacy such as thyme oil, eucalyptus oil, clove oil and the like, and a solubilizing or dispersing agent sufficient to form an aqueous solution or dispersion of said essential oils in a water carrier. Said compositions may further comprise other antimicrobial ingredients like phenolic compounds, quaternary ammonium compounds, however no betaine or sulphobetaine surfactants, or peroxygen bleaches are disclosed.

J-60038497 discloses a foam-generating two components detergent composition comprising (a) an aqueous hydrogen peroxide solution (0.5%-50%), (b) an alkaline compound containing an alkaline substance having 0.1% to 50% alkalinity expressed in terms of NaOH, like NaOH, KOH, Na2CO3. Also, at least one of components (a) and (b) comprises a surfactant and/or at least one compound (0.001% to 10%) selected from terpene alcohols, cyclic terpene alcohols and their esters, e.g., menthol, geraniol. This composition is intended to clean soils on hard materials like plastics, joints, and particularly difficult to clean recesses or corners. No reference is made to disinfection.

Summary of the invention

The present invention encompasses a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oils, or mixtures thereof, with the exception of aqueous compositions comprising from 0.5% to 50% by weight of the total composition of hydrogen peroxide, as said peroxygen bleach, and from 0.001% to 10% by

weight of a terpene alcohol, cyclic terpene alcohol or esters thereof, as the only antimicrobial active of essential oils.

The present invention further encompasses a process for disinfecting a surface wherein a liquid composition comprising a peroxygen bleach and an antimicrobial active of essential oil or a mixture thereof, is applied onto said surface.

The present invention also encompasses a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or mixtures thereof, packaged in a spray dispenser, as well as a wipe impregnated with a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or mixtures thereof.

Detailed description of the invention

The liquid disinfecting compositions according to the present invention comprise a peroxygen bleach and an antimicrobial active of essential oil, with the exception of aqueous compositions comprising from 0.5% to 50% by weight of the total composition of hydrogen peroxide and from 0.001% to 10% by weight of a terpene alcohol, cyclic terpene alcohol or ester thereof, as the only antimicrobial active of essential oils.

Accordingly, as an essential element the compositions according to the present invention comprise a peroxygen bleach, or mixtures thereof. Preferred peroxygen bleach is hydrogen peroxide, or a water soluble source thereof, or mixtures thereof. Hydrogen peroxide is most preferred to be used in the compositions according to the present invention.

It is believed that the presence of said peroxygen bleach especially hydrogen peroxide, persulfate and the like, in the compositions of the present invention contribute to the disinfection properties of said compositions. Indeed, said peroxygen bleach may attack the vital function of the micro-organism cells, for example, it may inhibit the assembling of ribosomes units within the cytoplasm of the micro-organism cells. Also, said peroxygen bleach like hydrogen peroxide, is a strong oxidizer that generates hydroxyl free radicals which attack proteins and nucleic acids.

Furthermore, the presence of said peroxygen bleach, especially hydrogen peroxide provides strong stain removal benefits which are particularly noticeable for example in laundry and hard surfaces applications.

As used herein a hydrogen peroxide source refers to any compound which produces hydrogen peroxide when said compound is in contact with water. Suitable water-soluble sources of hydrogen peroxide for use herein include percarbonates, persilicates, persulphates such as monopersulphate, perborates and peroxyacids such as diperoxydodecandioic acid (DPDA), magnesium perphthalic acid and mixtures thereof.

In addition, other classes of peroxides can be used as an alternative to hydrogen peroxide and sources thereof or in combination with hydrogen peroxide and sources thereof. Suitable classes include dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides and/or hydroperoxides.

Typically, the compositions herein comprise at least 0.01% by weight of the total composition of said peroxygen bleach, or mixtures thereof, preferably from 0.1% to 15%, and more preferably from 1% to 10%.

As a second essential ingredient, the compositions according to the present invention comprise an antimicrobial active of essential oil, or mixtures thereof. Typically, the compositions herein comprise at least 0.003% by weight of the total composition of said antimicrobial active of essential oil, or mixtures thereof, preferably from 0.006% to 10%, more preferably from 0.2% to 4% and most preferably from 0.2% to 2%.

Suitable antimicrobial actives of essential oils to be used in the compositions herein include any ingredient of essential oils that exhibit antimicrobial activity. It is speculated that said antimicrobial actives of essential oils act as proteins denaturing agents. Also, said antimicrobial actives of essential oils are compounds of natural origin which contribute to the safety profile of the compositions of the present invention when used to disinfect any surface. A further advantage of said actives of essential oils is that they may impart pleasant odor to the disinfecting compositions of the present invention without the need of adding a perfume. Indeed the disinfecting compositions

according to the present inv ntion deliver not only excellent disinfecting properties on clean surfaces to be disinfected but also good scent.
Such actives of essential oils include, but are not limited to, the antimicrobial actives obtained from oil of thyme, lemongrass, citrus, lemons, oranges, anise, clove, aniseed, cinnamon, geranium, roses, mint, lavender, citronella, eucalyptus, peppermint, camphor, sandalwood, cedar and mixtures thereof. Thus, actives of essential oils to be used herein include, but are not limited to, thymol (present for example in thyme oil), eugenol (present for example in cinnamon oil and clove oil), menthol (present for example in mint oil), geraniol (present for example in geranium oil and rose oil), verbenone (present for example in vervain oil), eucalyptol and pinocarvone (present for example in eucalyptus oil), cedrol (present for example in cedar oil), anethol (present for example in aniseed oil), carvacrol, hinokitiol, berberine, terpineol, limonene, ratanhiae and mixtures thereof. Preferred actives of essential oils to be used herein are thymol, eugenol, verbenone, eucalyptol, carvacrol, limonene and/or geraniol.

Thymol may be commercially available for example from Aldrich, eugenol may be commercially available for example from Sigma, Systems - Bioindustries (SBI) - Manheimer Inc.

It has now been found that a liquid composition comprising peroxygen bleach and an antimicrobial active of essential oils, or mixtures thereof exhibits improved chemical stability upon prolonged storage periods while delivering excellent disinfection performance on clean surfaces, even when used upon highly diluted conditions, as compared to the chemical stability obtained with the same composition comprising the corresponding antimicrobial essential oil instead of said antimicrobial active of essential oil. Indeed, the chemical decomposition of peroxygen bleach present in a liquid composition is reduced in the presence of said antimicrobial active of essential oil like eugenol, as compared to the presence of the corresponding essential oil like cinnamon oil and/or clove oil.

Chemical stability of the compositions herein may be evaluated by measuring the concentration of available oxygen (often abbreviated to Avox) at given storage time after having manufactured the compositions. The concentration of available oxygen can be measured by ch mical titration

methods known in the art, such as the iodimetric method, thiosulphatimetric method, the permanganometric method and the cerimetric method. Said methods and the criteria for the choice of the appropriate method are described for example in "Hydrogen Peroxide", W. C. Schumb, C. N. Satterfield and R. L. Wentworth, Reinhold Publishing Corporation, New York, 1955 and "Organic Peroxides", Daniel Swern, Editor Wiley Int. Science, 1970.

Excellent disinfection is obtained with the compositions of the present invention on a variety of micro-organisms including Gram positive bacteria like *Staphylococcus aureus*, and Gram negative bacteria like *Pseudomonas aeroginosa* as well as on fungi like *Candida albicans* present on clean surfaces, i.e., any surface being substantially free of organic and/or inorganic soils, even if used in highly diluted conditions.

Disinfection properties of a composition may be measured by the bactericidal activity of said composition. A test method suitable to evaluate the bactericidal activity of a composition on clean surfaces is described in European Standard, prEN 1040, CEN/TC 216 N 78, dated November 1995 issued by the European committee for standardisation, Brussels. European Standard, prEN 1040, CEN/TC 216 N 78, specifies a test method and requirements for the minimum bactericidal activity of a disinfecting composition. The test is passed if the bacterial colonies forming units (cfu) are reduced from a 10^7 cfu (initial level) to a 10^2 cfu (final level after contact with the disinfecting product), i.e. a 10^5 reduction of the viability is necessary. The compositions according to the present invention pass this test under clean conditions, even if used in highly diluted conditions.

Another test method suitable to evaluate the bactericidal activity of the present compositions on clean surfaces is AFNOR T72-190® and T72-301®

The compositions according to the present invention are aqueous liquid cleaning compositions. Said aqueous compositions have preferably a pH as is of not more than 12.0, more preferably from 2 to 6, and most

preferably from 3 to 5. The pH of the compositions can be adjusted by using organic acids like citric acid, succinic acid, acetic acid, aspartic acid, lactic acid and the like, or inorganic acids, or alkalinising agents.

The compositions of the present invention may further comprise surfactants known to those skilled in the art including nonionic, anionic, cationic, zwitterionic and/or amphoterics surfactants. Said surfactants are desirable as they contribute to the cleaning performance of the compositions herein.

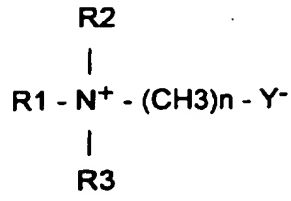
Typically, the compositions according to the present invention comprise up to 50% by weight of the total composition of a surfactant, or mixtures thereof, preferably from 0.01% to 30% and more preferably from 0.1% to 25%.

Accordingly, the compositions of the present invention may preferably comprise an amphoteric surfactant, or mixtures thereof. Suitable amphoteric surfactants to be used herein include betaine and sulphobetaine surfactants, derivatives thereof or mixtures thereof. Said betaine or sulphobetaine surfactants are preferred herein as they contribute to the disinfecting properties of the compositions herein. Indeed, they help disinfection by increasing the permeability of the bacterial cell wall, thus allowing other active ingredients to enter the cell.

Furthermore, due to the mild action profile of said betaine or sulphobetaine surfactants, the compositions herein comprising them may be particularly suitable for the cleaning of delicate surfaces, e.g. delicate laundry or surfaces in contact with food and/or babies. Betaine and sulphobetaine surfactants are also extremely mild to the skin and/or surfaces to be treated.

Suitable betaine and sulphobetaine surfactants to be used in the compositions of the present invention are the betaine/sulphobetaine and betaine-like detergents wherein the molecule contains both basic and acidic groups which form an inner salt giving the molecule both cationic and anionic hydrophilic groups over a broad range of pH values. Some common examples of these detergents are described in U.S. Pat. Nos. 2,082,275, 2,702,279 and 2,255,082, incorporated herein by reference.

Preferred betaine and sulphobetaine surfactants are according to the formula



wherein R1 is an alkyl radical containing from 1 to 24 carbon atoms, preferably from 8 to 18, and more preferably from 12 to 14, wherein R2 and R3 contain from 1 to 3 carbon atoms, and preferably 1 carbon atom, wherein n is an integer from 1 to 10, preferably from 1 to 6 and more preferably is 1, Y is selected from the group consisting of carboxyl and sulfonyl radicals and wherein the sum of R1, R2 and R3 radicals is from about 14 to about 24 carbon atoms, or mixtures thereof.

Examples of particularly suitable betaine surfactants include C12-C18 alkyl dimethyl betaine such as coconutbetaine and C10-C16 alkyl dimethyl betaine such as laurylbetaine.

Coconutbetaine is commercially available from Seppic under the trade name of Amonyl 265®. Laurylbetaine is commercially available from Albright & Wilson under the trade name Empigen BB/L®.

Other suitable amphoteric surfactants to be used herein include amine oxides or mixtures thereof. Amine oxides are preferred herein as they contribute to the disinfecting properties of the compositions herein. Indeed, they help disinfection by disrupting the cell wall/membrane of the bacteria, thus allowing other antimicrobial ingredients to enter the cell and for example attack the inner part of the cell.

Suitable amine oxides to be used herein have the following formula $R_1R_2R_3NO$ wherein each of R1, R2 and R3 is independently a saturated linear or branched hydrocarbon chain containing from 1 to 30 carbon atoms. Suitable amine oxides to be used according to the present invention are amine oxides having the following formula $R_1R_2R_3NO$ wherein R1 is a hydrocarbon chain containing from 1 to 30 carbon atoms, preferably from 6 to 20, mor preferably from 6 to 14 and most preferably

from 8 to 10, and wherein R2 and R3 are independently substituted or unsubstituted, linear or branched hydrocarbon chains containing from 1 to 4 carbon atoms, preferably of from 1 to 3 carbon atoms, and more preferably are methyl groups. R1 may be a saturated linear or branched hydrocarbon chain.

Preferred amine oxides for use herein are for instance natural blend C8-C10 amine oxides as well as C12-C16 amine oxides commercially available from Hoechst.

In a preferred embodiment of the present invention where the compositions herein are particularly suitable for the disinfection of a hard-surface, the surfactant is typically a surfactant system comprising an amine oxide and a betaine or sulphobetaine surfactant, preferably in a weight ratio of amine oxide to betaine or sulphobetaine of 2:1 to 100:1, more preferably of 6:1 to 100:1 and most preferably 10:1 to 50:1. The use of such a surfactant system in the compositions herein suitable for disinfecting a hard-surface, provides effective cleaning performance and provides shine on the cleaned surfaces, i.e., the amount of filming/streaking left on the cleaned surface that has been treated with said compositions is minimal.

The compositions herein may also preferably comprise an anionic surfactant or mixtures thereof.

Particularly suitable anionic surfactants to be used herein include water-soluble salts or acids of the formula $ROSO_3M$ wherein R is preferably a C_6-C_{24} hydrocarbyl, preferably an alkyl or hydroxyalkyl having a C_8-C_{20} alkyl component, more preferably a C_8-C_{16} alkyl or hydroxyalkyl, and M is H or a cation, e.g., an alkali metal cation (e.g., sodium, potassium, lithium), or ammonium or substituted ammonium (e.g., methyl-, dimethyl-, and trimethyl ammonium cations and quaternary ammonium cations, such as tetramethyl-ammonium and dimethyl piperdinium cations and quaternary ammonium cations derived from alkylamines such as ethylamine, diethylamine, triethylamine, and mixtures thereof, and the like).

Other suitable anionic surfactants to be used herein include alkyl-diphenyl-ether-sulphonates and alkyl-carboxylates. Other anionic surfactants can

include salts (including, for xample, sodium, potassium, ammonium, and substituted ammonium salts such as mono-, di- and triethanolamine salts) of soap, C_9-C_{20} linear alkylbenzenesulfonates, C_8-C_{22} primary or secondary alkanesulfonates, C_8-C_{24} olefinsulfonates, sulfonated polycarboxylic acids prepared by sulfonation of the pyrolyzed product of alkaline earth metal citrates, e.g., as described in British patent specification No. 1,082,179, C_8-C_{24} alkylpolyglycolethersulfates (containing up to 10 moles of ethylene oxide); alkyl ester sulfonates such as C_{14-16} methyl ester sulfonates; acyl glycerol sulfonates, fatty oleyl glycerol sulfates, alkyl phenol ethylene oxide ether sulfates, paraffin sulfonates, alkyl phosphates, isethionates such as the acyl isethionates, N-acyl taurates, alkyl succinamates and sulfosuccinates, monoesters of sulfosuccinate (especially saturated and unsaturated C_{12}-C_{18} monoesters) diesters of sulfosuccinate (especially saturated and unsaturated C_6-C_{14} diesters), acyl sarcosinates, sulfates of alkylpolysaccharides such as the sulfates of alkylpolyglucoside (the nonionic nonsulfated compounds being described below), branched primary alkyl sulfates, alkyl polyethoxy carboxylates such as those of the formula $RO(CH_2CH_2O)_kCH_2COO\text{-}M^+$ wherein R is a C_8-C_{22} alkyl, k is an integer from 0 to 10, and M is a soluble salt-forming cation. Resin acids and hydrogenated resin acids are also suitable, such as rosin, hydrogenated rosin, and resin acids and hydrogenated resin acids present in or derived from tall oil. Further examples are given in "Surface Active Agents and Detergents" (Vol. I and II by Schwartz, Perry and Berch). A variety of such surfactants are also generally disclosed in U.S. Patent 3,929,678, issued December 30, 1975 to Laughlin, et al. at Column 23, line 58 through Column 29, line 23.

Preferred anionic surfactants for use in the compositions herein are the C8-C16 alkyl sulfonates, C8-C16 alkyl sulfates, C8-C16 alkyl alkoxylated sulfates (e.g., C8-C16 alkyl ethoxylated sulfates), and mixtures thereof. Such anionic surfactants are preferred herein as it has been found that they contribute to the disinfecting properties of a disinfecting composition comprising a peroxygen bleach and/or an antimicrobial active of essential oil. For example, C8-C16 alkyl sulfate acts by disorganizing the bacteria cell menbrane, inhibiting enzymatic activities, interrupting the cellular transport and/or denaturing cellular proteins. Indeed, it is speculated that the improved disinfecting performance associated with th addition of an anionic

surfactant, especially a C8-C16 alkyl sulfonate, a C8-C16 alkyl sulfate and/or a C8-C16 alkyl alkoxylated sulfate, in for example a composition of the present invention, is likely due to multiple mode of attack of said surfactant against the bacteria. Thus, another aspect of the present invention is the use of an anionic surfactant, especially a C8-C16 alkyl sulfonate, a C8-C16 alkyl sulfate and/or a C8-C16 alkyl alkoxylated sulfate, in a disinfecting composition comprising a peroxygen bleach and/or an antimicrobial active of essential oil, to improve the disinfecting properties of said composition on gram negative and/or gram positive bacteria.

Suitable nonionic surfactants for use herein are fatty alcohol ethoxylates and/or propoxylates which are commercially available with a variety of fatty alcohol chain lengths and a variety of ethoxylation degrees. Indeed, the HLB values of such alkoxylated nonionic surfactants depend essentially on the chain length of the fatty alcohol, the nature of the alkoxylation and the degree of alkoxylation. Surfactant catalogues are available which list a number of surfactants, including nonionics, together with their respective HLB values.

Particularly suitable for use herein as nonionic surfactants are hydrophobic nonionic surfactants having an HLB (hydrophilic-lipophilic balance) below 16, preferably below 15 and more preferably below 14. Those hydrophobic nonionic surfactants have been found to provide good grease cutting properties.

Preferred hydrophobic nonionic surfactants to be used in the compositions according to the present invention are surfactants having an HLB below 16 and being according to the formula $RO-(C_2H_4O)_n(C_3H_6O)_mH$, wherein R is a C_6 to C_{22} alkyl chain or a C_6 to C_{28} alkyl benzene chain, and wherein n+m is from 0 to 20 and n is from 0 to 15 and m is from 0 to 20, preferably n+m is from 1 to 15 and, n and m are from 0.5 to 15, more preferably n+m is from 1 to 10 and, n and m are from 0 to 10. The preferred R chains for use herein are the C_8 to C_{22} alkyl chains. Accordingly, suitable hydrophobic nonionic surfactants for use herein are Dobanol ^{R} 91-2.5 (HLB= 8.1; R is a mixture of C9 and C_{11} alkyl chains, n is 2.5 and m is 0), or Lutensol ^{R} TO3 (HLB=8; R is a C_{13} alkyl chains, n is 3 and m is 0), or Lutensol ^{R} AO3 (HLB=8; R is a mixtur of C_{13} and C_{15}

alkyl chains, n is 3 and m is 0), or Tergitol ^{R} 25L3 (HLB= 7.7; R is in th range of C_{12} to C_{15} alkyl chain length, n is 3 and m is 0), or Dobanol ^{R} 23-3 (HLB=8.1; R is a mixture of C_{12} and C_{13} alkyl chains, n is 3 and m is 0), or Dobanol ^{R} 23-2 (HLB=6.2; R is a mixture of C_{12} and C_{13} alkyl chains, n is 2 and m is 0), or Dobanol ^{R} 45-7 (HLB=11.6; R is a mixture of C_{14} and C_{15} alkyl chains, n is 7 and m is 0) Dobanol ^{R} 23-6.5 (HLB=11.9; R is a mixture of C_{12} and C_{13} alkyl chains, n is 6.5 and m is 0), or Dobanol ^{R} 25-7 (HLB=12; R is a mixture of C_{12} and C_{15} alkyl chains, n is 7 and m is 0), or Dobanol ^{R} 91-5 (HLB=11.6; R is a mixture of C_9 and C_{11} alkyl chains, n is 5 and m is 0), or Dobanol ^{R} 91-6 (HLB=12.5 ; R is a mixture of C_9 and C_{11} alkyl chains, n is 6 and m is 0), or Dobanol ^{R} 91-8 (HLB=13.7 ; R is a mixture of C_9 and C_{11} alkyl chains, n is 8 and m is 0), Dobanol ^{R} 91-10 (HLB=14.2 ; R is a mixture of C_9 to C_{11} alkyl chains, n is 10 and m is 0), or mixtures thereof. Preferred herein are Dobanol ^{R} 91-2.5 , or Lutensol ^{R} TO3, or Lutensol ^{R} AO3, or Tergitol ^{R} 25L3, or Dobanol ^{R} 23-3, or Dobanol ^{R} 23-2, or mixtures thereof. These Dobanol^{R} surfactants are commercially available from SHELL. These Lutensol^{R} surfactants are commercially available from BASF and these Tergitol ^{R} surfactants are commercially available from UNION CARBIDE.

Other suitable surfactants also include C6-C20 conventional soaps (alkali metal salt of a C6-C20 fatty acid, preferably sodium salts).

The compositions according to the present invention may comprise as a preferred optional ingredient further antimicrobial ingredients that contribute to the antimicrobial activity of compositions of the present invention. Such ingredients include parabens like ethyl paraben, propyl paraben, methyl paraben, glutaraldehyde or mixtures thereof.

The compositions herein may further comprise a chelating agent as a preferred optional ingredient. Suitable chelating agents may be any of those known to those skilled in the art such as the ones selected from the group comprising phosphonat chelating agents, aminophosphonate

chelating agents, substituted heteroaromatic chelating agents, amino carboxylate chelating agents, other carboxylate chelating agents, polyfunctionally-substituted aromatic chelating agents, biodegradable chelating agents like ethylene diamine N,N'- disuccinic acid, or mixtures thereof.

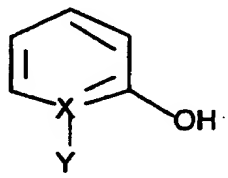
Suitable phosphonate chelating agents to be used herein include etidronic acid (1-hydroxyethylene-diphosphonic acid (HEDP)), and/or alkali metal ethane 1-hydroxydiphosphonates.

Suitable amino phosphonate chelating agents to be used herein include amino alkylene poly (alkylene phosphonates), nitrilotris(methylene)triphosphonates, ethylene diamine tetra methylene phosphonates, and/or diethylene triamine penta methylene phosphonates. Preferred aminophosphonate chelating agents to be used herein are diethylene triamine penta methylene phosphonates.

These phosphonate/amino phosphonate chelating agents may be present either in their acid form or as salts of different cations on some or all of their acid functionalities. Such phosphonate/amino phosphonate chelating agents are commercially available from Monsanto under the trade name DEQUEST®.

Substituted heteroaromatic chelating agents to be used herein include hydroxypiridine-N-oxide or a derivative thereof.

Suitable hydroxy pyridine N-oxides and derivatives thereof to be used according to the present invention are according to the following formula:



wherein X is nitrog n, Y is one of the following groups oxygen, -CHO, -OH, -(CH2)n-COOH, wherein n is an integer of from 0 to 20, preferably of from 0 to 10 and more preferably is 0, and wherein Y is preferably oxygen. Accordingly particularly preferred hydroxy pyridine N-oxides and derivatives thereof to be used herein is 2-hydroxy pyridine N-oxide.

Hydroxy pyridine N-oxides and derivatives thereof may be commercially available from Sigma.

Polyfunctionally-substituted aromatic chelating agents may also be useful in the compositions herein. See U.S. patent 3,812,044, issued May 21, 1974, to Connor et al. Preferred compounds of this type in acid form are dihydroxydisulfobenzenes such as 1,2-dihydroxy -3,5-disulfobenzene.

A preferred biodegradable chelating agent for use herein is ethylene diamine N,N'- disuccinic acid, or alkali metal, or alkaline earth, ammonium or substitutes ammonium salts thereof or mixtures thereof. Ethylenediamine N,N'- disuccinic acids, especially the (S,S) isomer have been extensively described in US patent 4, 704, 233, November 3, 1987 to Hartman and Perkins. Ethylenediamine N,N'- disuccinic acid is, for instance, commercially available under the tradename ssEDDS® from Palmer Research Laboratories. Ethylene diamine N,N'- disuccinic acid is particularly suitable to be used in the compositions of the present invention.

Suitable amino carboxylate chelating agents useful herein include ethylene diamine tetra acetates, diethylene triamine pentaacetates, diethylene triamine pentoacetate (DTPA), N-hydroxyethylethylenediamine triacetates, nitrilotri-acetates, ethylenediamine tetraproprionates, triethylenetetraaminehexa-acetates, ethanoldiglycines, propylene diamine tetracetic acid (PDTA) and methyl glycine di-acetic acid (MGDA), both in their acid form, or in their alkali metal, ammonium, and substituted ammonium salt forms. Particularly suitable to be used herein are diethylene triamine penta acetic acid (DTPA), propylene diamine tetracetic acid (PDTA) which is, for instance, commercially available from BASF under the trade name Trilon FS® and methyl glycine di-acetic acid (MGDA).

Further carboxylate chelating agents to be used herein includes malonic acid, salicylic acid, glycine, aspartic acid, glutamic acid, or mixtures thereof.

Said chelating agents, especially phosphonate chelating agents like diethylene triamine penta methylene phosphonates, are particularly preferred in the compositions according to the present invention as they have been found to further contribute to the disinfecting properties of hydrogen peroxide. Thus, another aspect of the present invention is the use of a chelating agent, especially a phosphonate chelating agent like diethylene triamine penta methylene phosphonate, in a disinfecting composition comprising hydrogen peroxide, to improve the disinfecting properties of said composition on gram negative and/or gram positive bacteria.

Typically, the compositions according to the present invention comprise up to 5% by weight of the total composition of a chelating agent, or mixtures thereof, preferably from 0.002% to 3% by weight and more preferably from 0.002% to 1.5%.

The compositions herein may comprise a radical scavenger as a preferred optional ingredient. Suitable radical scavengers for use herein include the well-known substituted mono and di hydroxy benzenes and derivatives thereof, alkyl- and aryl carboxylates and mixtures thereof. Preferred radical scavengers for use herein include di-tert-butyl hydroxy toluene (BHT), p-hydroxy-toluene, hydroquinone (HQ), di-tert-butyl hydroquinone (DTBHQ), mono-tert-butyl hydroquinone (MTBHQ), tert-butyl-hydroxy anysole (BHA), p-hydroxy-anysol, benzoic acid, 2,5-dihydroxy benzoic acid, 2,5-dihydroxyterephtalic acid, toluic acid, catechol, t-butyl catechol, 4-allyl-catechol, 4-acetyl catechol, 2-methoxy-phenol, 2-ethoxy-phenol, 2-methoxy-4-(2-propenyl)phenol, 3,4-dihydroxy benzaldehyde, 2,3-dihydroxy benzaldehyde, benzylamine, 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl) butane, tert-butyl-hydroxy-anyline, p-hydroxy anyline as well as n-propyl-gallate. Highly preferred for use herein are di-tert-butyl hydroxy toluene, which is for example commercially available from SHELL under the trade name IONOL CP® and/or tert-butyl-hydroxy anysole. These radical

scavengers further contribute to the stability of the peroxygen bleach-containing compositions herein.

Typically, the compositions according to the present invention comprise up to 5% by weight of the total composition of a radical scavenger, or mixtures thereof, preferably from 0.001% to 1.5% by weight and more preferably from 0.01% to 1%.

The compositions herein may comprise as a preferred optional ingredient a solvent or mixtures thereof. When used, solvents will, advantageously, give an enhanced cleaning to the compositions herein. Suitable solvents for incorporation in the compositions according to the present invention include propylene glycol derivatives such as n-butoxypropanol or n-butoxypropoxypropanol, water-soluble CARBITOL® solvents or water-soluble CELLOSOLVE® solvents. Water-soluble CARBITOL® solvents are compounds of the 2-(2-alkoxyethoxy)ethanol class wherein the alkoxy group is derived from ethyl, propyl or butyl. A preferred water-soluble carbitol is 2-(2-butoxyethoxy)ethanol also known as butyl carbitol. Water-soluble CELLOSOLVE® solvents are compounds of the 2-alkoxyethoxyethanol class, with 2-butoxyethoxyethanol being preferred. Other suitable solvents are benzyl alcohol, methanol, ethanol, isopropyl alcohol and diols such as 2-ethyl-1,3-hexanediol and 2,2,4-trimethyl-1,3-pentanediol and mixture thereof. Preferred solvents for use herein are n-butoxypropoxypropanol, butyl carbitol® benzyl alcohol, isopropanol and mixtures thereof. Most preferred solvents for use herein are butyl carbitol®, benzyl alcohol and isopropanol.

The solvents may typically be present within the compositions of the invention at a level up to 15% by weight, preferably from 2% to 7% by weight of the composition.

The compositions herein may further comprise a variety of other optional ingredients such as buffers (e.g. borate buffers), builders, stabilisers, bleach activators, soil suspenders, dye transfer agents, brighteners, perfumes, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, p rfumes and dy s.

Packaging form of the compositions:

The compositions herein may be packaged in a variety of suitable detergent packaging known to those skilled in the art. The liquid compositions herein may desirably be packaged in manually operated spray dispensing containers, which are usually made of synthetic organic polymeric plastic materials. Accordingly, the present invention also encompasses liquid disinfecting compositions comprising a peroxygen bleach and an antimicrobial active of essential oil packaged in a spray dispenser, preferably in a trigger spray dispenser or a pump spray dispenser. These liquid compositions may further comprise optional ingredients as specified herein before.

Indeed, said spray-type dispensers allow to uniformly apply to a relatively large area of a surface to be disinfected a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, thereby contributing to disinfection properties of said composition. Such spray-type dispensers are particularly suitable to disinfect vertical surfaces.

Suitable spray-type dispensers to be used according to the present invention include manually operated foam trigger-type dispensers sold for example by Specialty Packaging Products, Inc. or Continental Sprayers, Inc. These types of dispensers are disclosed, for instance, in US-4,701,311 to Dunnining et al. and US-4,646,973 and US-4,538,745 both to Focarracci. Particularly preferred to be used herein are spray-type dispensers such as T 8500® or T 8900® commercially available from Continental Spray International or T 8100® commercially available from Canyon, Northern Ireland. In such a dispenser the liquid composition is divided in fine liquid droplets resulting in a spray that is directed onto the surface to be treated. Indeed, in such a spray-type dispenser the composition contained in the body of said dispenser is directed through the spray-type dispenser head via energy communicated to a pumping mechanism by the user as said user activates said pumping mechanism. More particularly, in said spray-type dispenser head the composition is forced against an obstacle, e.g. a grid or a cone or the like, thereby providing shocks to help atomise the liquid composition, i.e. to help the formation of liquid dropl ts.

The present invention also encompasses disinfecting wipes. By "wipes" it is meant herein disposable towels, e.g. disposable paper towels, impregnated with a liquid composition comprising a peroxygen bleach and an antimicrobial active of essential oil, e.g. said wipes may be wetted with said composition. These liquid compositions may further comprise optional ingredients as specified herein before.

Preferably said wipes are packaged in a plastic box. The advantage of this execution is a faster usage of a disinfecting composition by the user, this even outside the house, i.e. there is no need to pour the liquid compositions according to the present invention on the surfaces to be treated/disinfect and to dry it out with a cloth. In other words, wipes allow disinfection of surfaces in one step.

The process of disinfecting:

In its broadest aspect, the present invention encompasses a process of disinfecting surfaces wherein the composition used is a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oils, or mixtures thereof.

By "surface" it is meant herein any surface including animate surface like human skin, mouth, teeth, and inanimate surfaces. Inanimate surfaces include, but are not limited to, hard-surfaces typically found in houses like kitchens, bathrooms, or in car interiors, e.g., tiles, walls, floors, chrome, glass, smooth vinyl, any plastic, plastified wood, table top, sinks, cooker tops, dishes, sanitary fittings such as sinks, showers, shower curtains, wash basins, WCs and the like, as well as fabrics including clothes, curtains, drapes, bed linens, bath linens, table cloths, sleeping bags, tents, upholstered furniture and the like, and carpets. Inanimate surfaces also include household appliances including, but not limited to, refrigerators,

freezers, washing machines, automatic dryers, ovens, microwave ovens, dishwashers and so on.

In the process of disinfecting surfaces according to the present invention said liquid compositions may be applied to the surface to be disinfected in its neat form or in its diluted form.

By "diluted form" it is meant herein that the liquid compositions to be used in the disinfection process herein may be diluted by the user typically up to 100 times their weight of water, preferably into 80 to 30 times their weight of water, and more preferably 60 to 40 times.

In the preferred embodiment of the process of the present invention wherein said liquid composition is applied to a hard-surface to be disinfected in its diluted form, it is not necessary to rinse the surface after the composition has been applied, indeed no visible residues are left onto the surface.

The present invention will be further illustrated by the following examples.

Examples

The following compositions were made by mixing the listed ingredients in the listed proportions (weight % unless otherwise specified). These compositions passed the prEN 1040 test of the European committee of standardisation. These compositions provide excellent disinfection when used neat or diluted, e.g. at 1:100, 1:25, 1:50 dilution levels, on clean surfaces while delivering also excellent surface safety and exhibiting excellent stability upon prolonged storage periods.

Compositions	I	II	III	IV	V	VI

Hydrogen peroxide	7.0	6.0	--	6.0	6.0	2.0
Monopersulfate	--	--	2.0	--	--	--
Thymol	0.5	--	--	--	0.5	--
Eugenol	--	1.0	--	--	--	0.1
Geraniol	0.5		1.5	--	--	--
Eucalyptol	--	--	--	1.0	--	--
Water and minors	up to 100%					
--						
H2SO4 up to pH 4						

Compositions	VII	VIII	IX	X
Hydrogen peroxide	7.0	6.0	--	1.0
Monopersulfate	--	--	2.0	--
Thymol	--	--	--	--
Eugenol/eucalyptol (1:1)	1.0	--	0.5	0.05
Geraniol	--	0.5	--	--
Eucalyptol	--	0.5	--	--
Water and minors	up to 100%			
H2SO4 up to pH 4				

Compositions	XI	XII	XIII	XIV	XV
Hydrogen peroxide	7.0	6.0	--	1.0	1.0
Monopersulfate	--	--	2.0	--	--
C10 Alkyl Sulphate	4.0	3.0	1.5	0.9	--
C10 Amine Oxide	--	1.5	0.5	0.6	0.9
Lauryl betaine	--	--	--	--	0.05
Thymol	--	--	--	--	0.03
Eugenol/eucalyptol (1:1)	1.0	--	0.5	0.05	--
Geraniol	--	0.5	--	--	--
Eucalyptol	--	0.5	--	--	0.02
Water and minors	up to 100%				
H2SO4 up to pH 4					

WHAT IS CLAIMED IS:

1. A liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or mixtures thereof, with the exception of aqueous compositions comprising from 0.5% to 50% by weight of the total composition of hydrogen peroxide, as said peroxygen bleach, and from 0.001% to 10% by weight of a terpene alcohol, cyclic terpene alcohol or ester thereof, as the only antimicrobial active of essential oils.

2. A process of disinfecting a surface wherein a liquid composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or a mixture thereof, is applied onto said surface.

3. A process according to claim 2 wherein said composition is aqueous and is applied onto said surface after having been diluted up to 100 times its weight of water, preferably into 80 to 40 times its weight of water, and more preferably 60 to 30 times.

4. A composition or process according to any of the preceding claims wherein said peroxygen bleach is hydrogen peroxide or a water soluble source thereof selected from the group consisting of percarbonates, persilicates, persulphates, perborates, peroxyacids, dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides, organic and inorganic hydroperoxides and mixtures thereof, and preferably is hydrogen peroxide .

5. A composition or process according to any of the preceding claims wherein said composition comprises at least 0.01% by weight of the total composition of said peroxygen bleach, or mixtures thereof, pr ferably from 0.1% to 15% and more preferably from 1% to 10%.

6. A composition or process according to any of the preceding claims wherein said antimicrobial active of essential oil is thymol, eugenol, menthol, geraniol, verbenone, eucalyptol, pinocarvone, cedrol, carvacrol, anethol, hinokitiol, berberine, terpineol, limonene, ratanhiae or mixtures thereof and preferably is thymol, eugenol, verbenone, eucalyptol, carvacrol, limonene and/or geraniol.

7. A composition or process according to any of the preceding claims wherein said composition comprises at least 0.003% by weight of the total composition of said antimicrobial active of essential oil, or mixtures thereof, preferably from 0.006% to 10% and more preferably from 0.2% to 4%.

8. A composition or process according to any of the preceding claims wherein said composition further comprises a surfactant selected from the group consisting of anionic surfactants, nonionic surfactants, cationic surfactants, amphoteric surfactants, zwitterionic surfactants and mixtures thereof, and wherein said surfactant is present up to a level of 50% by weight of the total composition, preferably at a level of from 0.01% to 30%, and more preferably of from 0.1% to 25%.

9. A composition or process according to claim 8 wherein said surfactant is an amphoteric surfactant or a mixture thereof, preferably a betaine or sulphobetaine surfactant, or a derivative thereof, or a mixture thereof according to the following formula

$$R1 - \overset{\displaystyle R2}{\underset{\displaystyle R3}{N^{+}}} - (CH3)n - Y^{-}$$

wherein R1 is an alkyl radical containing from 1 to 24 carbon atoms, preferably from 8 to 18, and more preferably from 12 to 14, wherein R2 and R3 contain from 1 to 3 carbon atoms, and preferably 1 carbon atom, wh r in n is an integ r from 1 to 10, preferably from 1 to 6 and

more preferably is 1, Y is selected from the group consisting of carboxyl and sulfonyl radicals and wherein the sum of R1, R2 and R3 radicals is from about 14 to about 24 carbon atoms and/or an amine oxide according to the formula $R_1R_2R_3NO$ wherein each of R1, R2 and R3 is independently a saturated linear or branched hydrocarbon chain containing from 1 to 30 carbon atoms, preferably from 6 to 20, and wherein R2 and R3 are independently substituted or unsubstituted, linear or branched hydrocarbon chains containing from 1 to 4 carbon atoms, preferably of from 1 to 3 carbon atoms, and more preferably are methyl groups.

10. A composition or process according to any of the preceding claims wherein said composition further comprises a chelating agent selected from the group consisting of phosphonate chelating agents, aminophosphonate chelating agents, substituted heteroaromatic chelating agents, amino carboxylate chelating agents, other carboxylate chelating agents, polyfunctionally-substituted aromatic chelating agents, ethylene diamine N,N'- disuccinic acid, and mixtures thereof.

11. A composition or process according to any of the preceding claims wherein said composition further comprises at least an optional ingredient selected from the group consisting of radical scavengers, solvents, buffers, builders, stabilisers, bleach activators, soil suspenders, dye transfer agents, brighteners, perfumes, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, perfumes, dyes and mixtures thereof.

12. A composition or process according to any of the preceding claims wherein said composition further comprises another antimicrobial ingredient selected from the group consisting of glutaraldehyde, ethyl paraben, propyl paraben, methyl paraben and mixtures thereof.

13. A composition or process according to any of the preceding claims wherein said composition has of a pH of no more than 12, preferably below 7, more preferably of from 2 to 6, and most preferably of from 3 to 5.

14. A liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or mixtures thereof, packaged in a spray dispenser.

15. A wipe impregnated with a liquid disinfecting composition comprising a peroxygen bleach and an antimicrobial active of essential oil, or mixtures thereof.

16. A liquid composition according to claim 14 or a wipe according to claim 15 wherein said liquid composition comprises at least 0.01% by weight of the total composition of said peroxygen bleach, or mixtures thereof, preferably from 0.1% to 15% and more preferably from 1% to 10% and wherein said peroxygen bleach is hydrogen peroxide or a water soluble source thereof selected from the group consisting of percarbonates, persilicates, persulphates, perborates, peroxyacids, dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides, organic and inorganic hydroperoxides and mixtures thereof.

17. A liquid composition according to any of the preceding claims 14 or 16, or a wipe according to any of the claims 15 or 16 wherein said liquid composition comprises at least 0.003% by weight of the total composition of said antimicrobial active of essential oil, or mixtures thereof, preferably from 0.006% to 10% and more preferably from 0.2% to 4% and wherein said antimicrobial active of essential oil is thymol, eugenol, menthol, geraniol, verbenone, eucalyptol, pinocarvone, cedrol, carvacrol, anethol, hinokitiol, berberine, terpineol, limonene, ratanhiae or mixtures thereof.

18. The use of a chelating agent, preferably of a phosphonate chelating agent, more preferably of diethylene triamine penta methylene phosphonate, in a p roxygen bleach-containing composition to

improve th disinfecting properties of said composition on gram negative and/or gram positive bacteria.

19. The use of an anionic surfactant, especially a C8-C16 alkyl sulfonate, a C8-C16 alkyl sulfate and/or a C8-C16 alkyl alkoxylated sulfate, in a disinfecting composition comprising a peroxygen bleach and/or an antimicrobial active of essential oil, to improve the disinfecting properties of said composition on gram negative and/or gram positive bacteria.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/00327

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : A62D 3/00
US CL : 252/ 186.42,186.43; 510/370,372,375,378; 8/111; 424/195.1
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 252/ 186.42,186.43; 510/370,372,375,378; 8/111; 424/195.1

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

APS ESSENTIAL OIL# AND PEROXIDE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y,P	US 5,545,374 A (FRENCH ET AL.) 13 August 1996 See abstract and col 6 and 7	1-4,14-17
Y	US 5,403,587 A (McCUE ET AL.) 04 April 1995, col 3 line 1-27, col 4 line 19-29.	1-4,14-17, 19
Y	US 3,912,666 A (SPITZER ET AL.) 14 October 1975 See abstract and column 12 line 58 to column 13 line 33	14-17
X	US 4,767,617 A (SHANSKY ET AL.) 30 August 1988, See example 3 in column 4 lines 42-65	1,4
X	US 5,362,495 A (LESAGE) 08 November 1994, See claim 19	1,4
X	US 4,430,236 A (FRANKS) 07 February 1984, column 5 lines 5-35	1-4

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search
17 MARCH 1997

Date of mailing of the international search report
14 MAY 1997

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Form PCT/ISA/210 (second sheet)(July 1992)*

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/00327

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,347,149 A (SMITH ET AL.) 31 August 1982, see abstract	18
X	JP 60-038479 A (LION CORP.) 28 February 1985, see abstract	1-4

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US97/00327

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☒ Claims Nos.: 5-13,
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.